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Final Report

Sources and Chronology of Mercury Contamination in Cottage
Grove

Reservoir

For

U.S. Army Corps of Engineers

Portland, OR

May 20, 2003

From

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MERCURY DISTRIBUTION IN SEDIMENTS AND UPTAKE INTO AN AQUATIC FOOD WEB AT COTTAGE GROVE RESERVOIR, OREGON

INTRODUCTION

This work extends previous research (Park and Curtis 1997) at Cottage Grove Reservoir, located ten kilometers south of Cottage Grove, Oregon. This study examined mercury contamination in soils of the suspected point source and down-gradient tributary stream and reservoir sediments. Reservoir sediment core stratigraphy samples were also analyzed for mercury and assessed how contaminant loading changed over time. Analysis of core samples for ^{137}Cs and ^{210}Pb estimated sedimentation rates, and contributed to assessment of Black Butte Mine as a source of contamination to the reservoir over time. Mercury distribution in tributary stream sediments and reservoir sediment core stratigraphy supports the conclusion that Black Butte Mine is a point source of mercury contamination to the Cottage Grove Reservoir. Mercury concentrations in invertebrates and largemouth bass from the reservoir provided insight into food web contamination. Mercury concentrations in largemouth bass exceeding the State and Federal Action Limits.

The Cottage Grove Reservoir is a US Army Corps of Engineers flood-control reservoir located 10 kilometers south of the town of Cottage Grove, Oregon, near the southern end of the Willamette Valley in the Western Cascade Mountains (Fig. 1). The reservoir was constructed in 1942, with intent to regulate the flow of the headwaters of the Coast Fork of the Willamette River. The reservoir is seasonally managed for flood control, conservation storage, and water release to downstream areas. Cottage Grove Reservoir is located within the Willamette/Sandy basin and its watershed encompasses 257 square kilometers of land. The beneficial uses of the Cottage Grove Reservoir include resident fish and aquatic life, water contact recreation, fishing, and aesthetics. Most of the water rights within this watershed

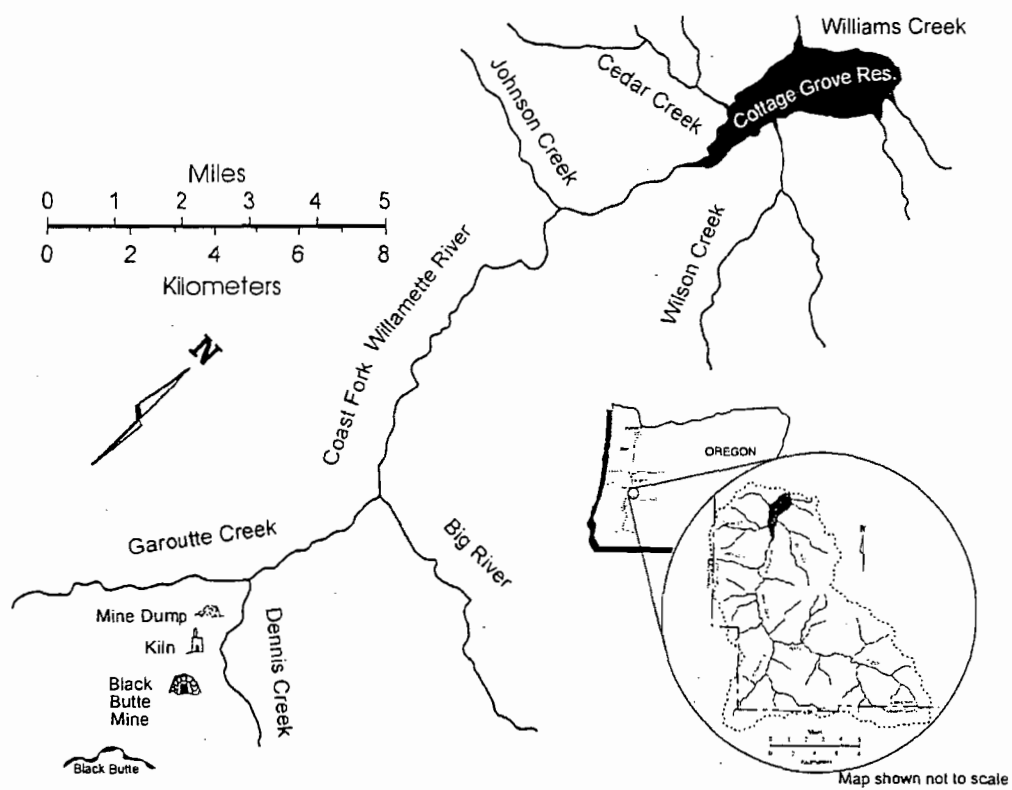


Figure 1: Cottage Grove Reservoir Site Map

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are for irrigation use, both on the Coast Fork and for miscellaneous streams (BLM, 1997).

Black Butte Mine is located approximately 13 kilometers south of the reservoir, and was once one of the largest producers of mercury in the United States. Mining in this area exploited Eocene marine sediments and volcanics from the Fisher Formation, where the mine yielded cinnabar ore. It was discovered in 1890 and operated intermittently until the early 1970s when the land containing the mine was sold for its timber assets (Orr et al. 1992). Sulfur, combined with the mercury was burned off in a 40-ton -a-day furnace. Mercury was produced from the Black Butte Mine from approximately 1882-1926, 1927 to 1943, 1951, 1957 to 1958, and 1965 to 1967; a total of 18,156 flasks of mercury were produced during that time (Brooks, 1971). Currently, it is estimated that 300,000 cubic yards of mine tailings remain in the vicinity of Black Butte mine and along Dennis Creek (BLM, 1997).

In western Oregon, cinnabar or mercury ore occurs scattered within a belt 20 miles wide that extends from Lane, Douglas, and Jackson counties in the southern Coast Range to the California border. In Lane County, the Black Butte and Bonanza mines are responsible for about one-half of Oregon's mercury production (Orr et al. 1992). Mercury amalgamation was also used in historic gold and silver mining operations around the state, so placer mining operations are also potential sources of contamination (Park and Curtis 1997; Bretagne et al. 2001).

Atmospheric transport and deposition of mercury is another important source of mercury, especially in remote and semi-remote areas. Mercury in air emissions is contributed from coal burning power plants, municipal waste incinerators, and other industrial sources.

The Cottage Grove Reservoir watershed is considered a point-source impacted water system as a result of historical mercury mining and processing within its watershed (Park and Curtis, 1997). Mercury enters the environment from ore

wastes and via atmospheric deposition of mercury vapor that escapes condensers during roasting of cinnabar (Bargagli, 1990). Although the watershed is likely to be influenced by the atmospheric deposition of mercury, mobilization of natural deposits, and small scale uses of the metal as an amalgamating agent in gold and silver mining, we hypothesized elevated concentrations found in this watershed primarily from past cinnabar mining and roasting activities at Black Butte Mine.

The general objective of this study is to assess a potential point source and determine the distribution of mercury contamination in Cottage Grove Reservoir and its tributary streams. Accomplishing the following explanatory objectives will address this general objective.

- Determine mercury concentrations in soils, and mine tailings on the Black Butte Mine site and Cottage Grove Reservoir tributary streams.
- Compare and contrast mercury stratigraphy in reservoir sediment cores from 1995 and 2002.
- Estimate sediment deposition rates and long-term trends for mercury accumulation in the reservoir.
- Assess mercury contamination in lower trophic levels in the aquatic food web from the reservoir.

MATERIALS AND METHODS

Two sediment cores, six surface sediment samples, and food web samples representing three trophic levels were collected from the Cottage Grove Reservoir. In addition, 26 surface sediment grab samples were collected from several of the tributaries throughout the watershed. Tributary and mine site sample locations are presented in Figure 2. With the exception of the mine site samples and one sediment core, all samples were collected between July and September 2002 and all samples collected were analyzed for total mercury. Mine site samples were collected during 1995 and analyzed for total mercury.

SAMPLE COLLECTION METHODS

Two sediment cores were collected from the deepest areas of the reservoir in 2002; the depth of the water at the collection point was approximately 16 meters. One sediment core was collected in the same area in 1995. Cores were collected by boat using a coring device with a detachable, 83 mm diameter PVC barrel. Cores obtained in 2002 were 36 cm in length, however the cores do not represent the complete thickness of lake sediment because of the absence of parent material from the bottom of each core (i.e. river gravel or sand). The core collected in 1995 was 24 cm. Following collection, each core was immediately cut into 2-cm intervals (resulting in 18 or 12 samples per core), placed in pretreated ICHÉM® glass jars, and placed in a cooler on ice until they reached the laboratory.

Six surface sediment samples were collected, representing a longitudinal transect through the center of the reservoir. Each surface sediment sample was collected by boat, using a ponar dredge at approximately one-half mile sampling intervals. Chironomid larvae samples were also collected by boat using the ponar dredge from locations near the spillway of the dam (where the sediment layer was estimated to be deepest). Surface sediment samples were collected and sieved

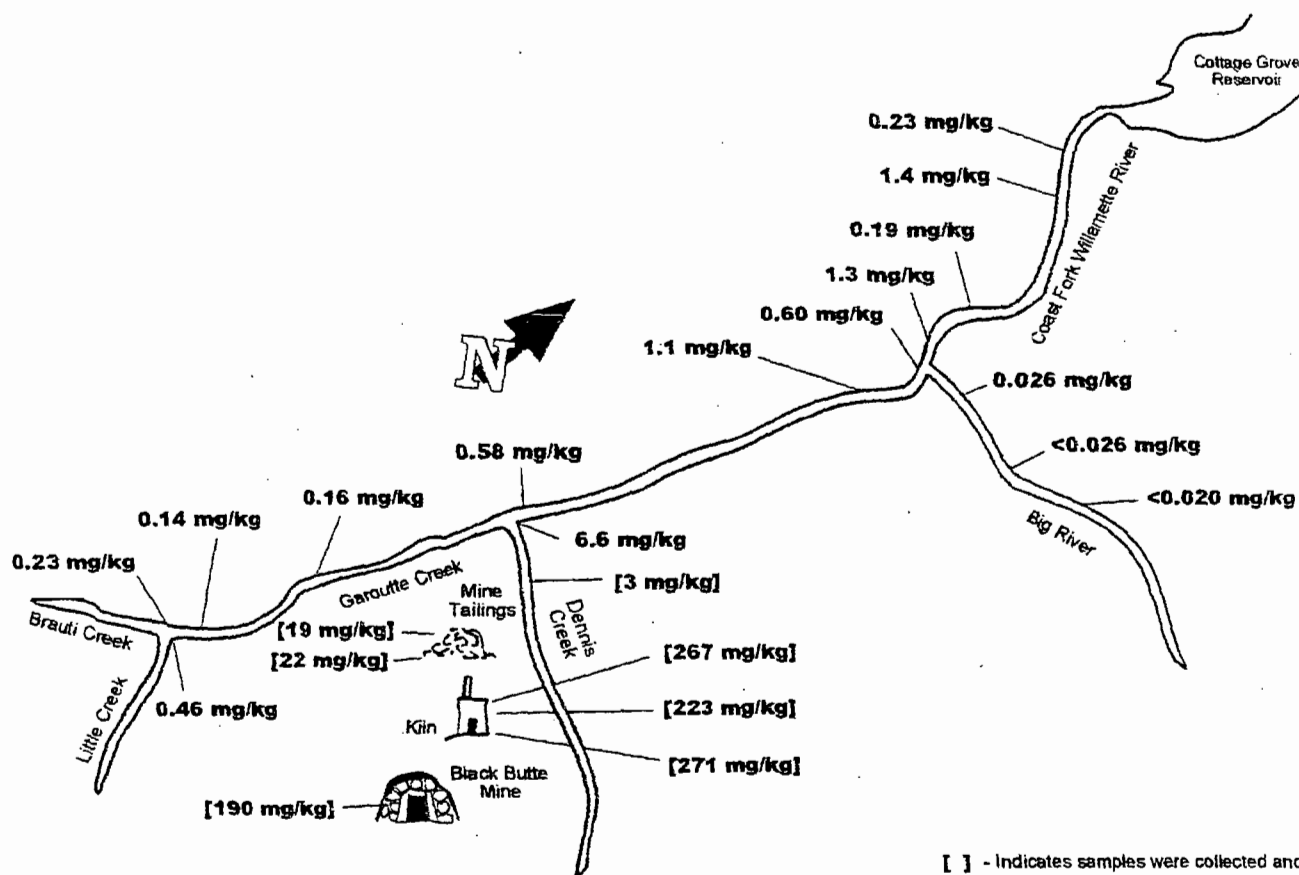


Figure 2: Mercury Concentrations Near Black Butte Mine and Surrounding Tributaries

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until the number of Chironomid larvae was sufficient for approximately three sample replicates.

Surface sediment grab samples were collected from each of the identified tributaries within the watershed. Particle size for each grab sample varied, and ranged from fine particulates to gravel material.

With the exception of largemouth bass tissue samples¹, all foodweb samples were collected from the reservoir during August and September 2002. Fingerling brown bullhead catfish, snails, bullfrog tadpoles, and Anisoptera and Zygoptera nymphs were collected from the reservoir using a sweepnet. With the exception of the Zygoptera nymphs, enough sample quantity was collected for three sample replicates. For the Zygoptera nymphs, only enough sample was collected for two sample replicates. Largemouth bass tissue data collected in June 1998 were provided by the Oregon Department of Environmental Quality (unpublished data).

All surface sediment, sediment grab samples, and foodweb samples were placed in pretreated ICHÉM® glass jars, and placed in a cooler on ice until they reached the laboratory. With the exception of the foodweb samples, all sediment samples were held in a cooler at 4°C until analysis. All foodweb samples were frozen until the time of analysis.

ANALYTICAL METHODS

Total mercury

All sediment and food web samples were analyzed for total mercury concentrations on a wet-weight basis in accordance with EPA Method 7471 (EPA, 1996) using a mercury

¹ Largemouth bass samples were not collected during 2002. Largemouth bass tissue samples were collected during 1998 and provided for use in this study by Eugene Foster of the Oregon Department of Environmental Quality (unpublished data).

autoanalyzer (Leeman Labs PS200). EPA Method 7471 determines total mercury concentrations using cold vapor atomic absorption.

Approximately 0.5 gram of sample was weighed into a clean BOD bottle, followed by the addition of 5 ml of concentrated sulfuric acid (H_2SO_4), 2 ml of concentrated nitric acid (HNO_3), and 5 ml of potassium permanganate (KMnO_4). Bottles were covered with aluminum foil and digested in an autoclave at 121°C for 15 minutes. After samples cooled, 6 ml of sodium chloride-hydroxylamine hydrochloride was dispensed into each bottle and the volume brought to 100 ml with ultra-pure water.

The instrument was calibrated based on a linear six-point calibration curve (zero, 0.1 ppb, 0.5 ppb, 1 ppb, 2 ppb, and 5 ppb), the linearity of each calibration curve was greater than 0.995. To monitor the calibration curve, a continuing calibration verification standard and blank sample was analyzed at a 10 percent frequency and at the end of each analytical batch. To verify the quality of the analytical results obtained, a standard reference material sample and matrix spike/matrix spike duplicates were analyzed with each analytical batch of 20 samples or less. All sample analyses were performed in duplicate by the instrument, with the average of the analyses reported. The method detection limit achieved was approximately 0.02 mg/kg. Percent total solid results were used to convert all wet-weight analysis results for sediment samples to a dry-weight basis.

Total solids

The percent total solids was determined for all sediment samples analyzed in accordance with EPA Method 160.3 (EPA, 1983). Subsamples were weighed in aluminum pans and placed in a drying oven at 115°F for 8 h. Samples were then cooled in a desiccator and weighed.

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Core dating

Select intervals within each 2002 sediment core were analyzed for excess ^{210}Pb , ^{226}Ra , and ^{137}Cs activity to estimate and sediment accumulation rates ages. The following sample intervals were selected for radioassay: 0-2 cm, 8-10 cm, 16-18 cm, 22-24 cm, 28-30 cm, and 34-36 cm. One-gram subsamples of dried sediment from each interval were submitted to the University of Liverpool Environmental Radiometric Laboratory.

^{210}Pb , ^{226}Ra , and ^{137}Cs were measured by direct gamma assay, using Ortec HPGe GWL series well-type coaxial low background intrinsic detectors (Appleby et al. 1986; Appleby et al. 1992) and dates were determined according to the c.r.s. (constant rate of supply) model (Appleby and Oldfield, 1978). A narrative interpretation of results including assessment of dating uncertainty was provided for each core dated.

Statistical analysis

Means and standard errors were calculated for the mercury analyses obtained from the set of lake-sediment cores and tributary surface sediment samples.

RESULTS

MINE SITE AND TRIBUTARY STREAM SAMPLES

Mercury concentrations were measured in six composite (5 subsamples per composite) surface soil samples and one surface sediment sample collected in 1995 (Fig. 2).

Mercury concentrations in surface soil were measured at locations near Black Butte Mine, the abandoned kiln, and locations surrounding the mine tailings. One surface sediment sample was collected from Dennis Creek. Elevated mercury concentrations were found in the samples collected at and near the mine. The mercury concentration measured at the mine was 190 mg/kg, concentrations surrounding the kiln ranged from 223 mg/kg to 271 mg/kg, and concentrations near the mine tailings were approximately 20 mg/kg.

Sediment from Dennis Creek, located directly adjacent to the mine and mine dumps areas, was sampled in 1995 and mercury was detected at 3 ppm (Fig. 2). Access was restricted in this area in 2002 and it was not sampled. One sample was collected from Dennis Creek immediately upstream of the confluence with Garoutte Creek in 2002 with a mercury concentration of 6.6 mg/kg (Fig. 2). Garoutte Creek sediments upstream of the confluence of Dennis Creek were also sampled and were much lower (Fig. 2).

Mercury concentrations were measured in surficial sediments/fines collected along the course of the Coast Fork of the Willamette River and another major tributary, Big River (Fig. 2). Three samples were collected at locations along Big River, upstream of the confluence with Garoutte and Dennis Creeks. Mercury concentrations along Big River were less than or equal to 0.02 mg/kg. Six samples were collected along Garoutte Creek at locations that are representative of the headwaters (0.14 mg/kg to 0.16 mg/kg), below the confluence with Dennis Creek (0.58 mg/kg), above the confluence with Big River (1.1 mg/kg and 1.3 mg/kg), and finally at the confluence with the Coast

Fork of the Willamette River (0.6 mg/kg). In addition, one sample was collected from Little Creek (0.46 mg/kg) and one from Brauti Creek (0.23 mg/kg), both of which form the head waters of Garoutte Creek. Samples (two per creek) were collected from smaller tributaries (Williams Creek, Wilson Creek, and Cedar Creek) that enter the reservoir directly and in the Coast Fork downstream of the reservoir (Fig. 3). Average mercury concentrations detected in Wilson, Cedar, and Williams Creek were 0.03 mg/kg, 0.06 mg/kg, and 0.07 mg/kg, respectively.

Four samples were collected downstream of the reservoir along the Coast Fork of the Willamette River. Mercury concentrations in downstream samples ranged from less than 0.02 mg/kg to 0.07 mg/kg.

TRANSECT SAMPLES

Mercury concentrations were measured in a transect of six reservoir surface sediment samples collected during 2002 (Fig. 3). Samples represent surface sediment mercury from the north end of the reservoir (near the dam) to the south end near the inlet of Coast Fork. Mercury concentrations ranged from 0.7 mg/kg near the inlet to a maximum of 3.6 mg/kg near the dam.

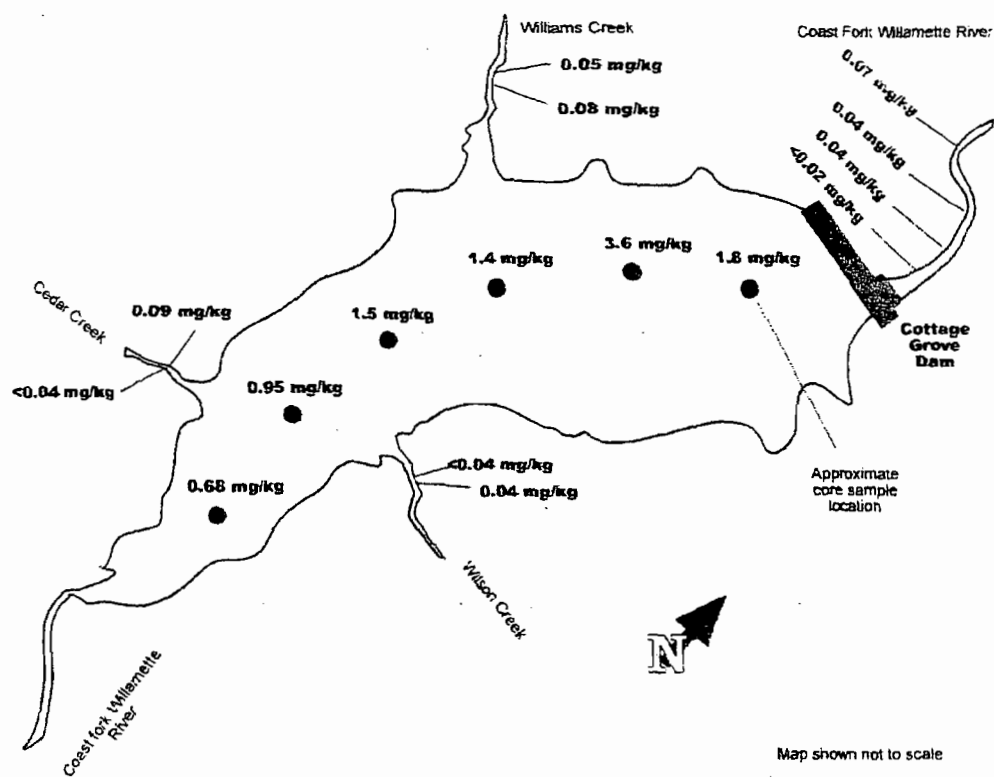


Figure 3: Mercury Concentrations in Transect and Tributary Surface Samples
(results are presented on dry-weight basis)

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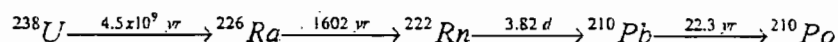
SEDIMENT CORES

Mean mercury concentrations were measured in 2 cm layers from the two cores collected from the reservoir in 2002 and one core collected in 1995. Mercury concentrations measured at each depth interval for each individual core sample from 2002 and the average and standard errors are presented in Table 1. Results for the core collected in 1995 were total mercury concentrations for each 2 cm layer (Table 2).

The pattern of mercury concentrations observed in the top 25 centimeters of 2002 and 1995 are generally consistent with one another. Concentrations of mercury from all core samples range from 0.7 mg/kg to 1.4 mg/kg at the surface. Mercury concentrations generally remain consistent or slightly decrease over the top 20 cm, with concentrations ranging from 0.5 mg/kg (at 12-14 cm) to 1.9 mg/kg (at 2-4 cm), with an average concentration of 0.9 mg/kg. Mercury concentrations generally increase over the last half of the core, with concentrations ranging from 0.8 mg/kg (20-24 cm) to 3.7 mg/kg (32-34 cm), with an average concentration of 2.0 mg/kg (Fig. 4).

Sediment core dating results and geochronology

^{210}Pb occurs naturally as one of the radionuclides in the ^{238}U decay series. The following shows the primary decay products and their half-lives in the ^{238}U decay series.



^{210}Pb is present in the atmosphere as an intermediate product of the gaseous isotope ^{222}Rn . ^{210}Pb is subsequently removed from the atmosphere as a result of rainfall or dry deposition, where it then falls to land surface. ^{210}Pb that falls into the water column is scavenged and deposited to the bed of the reservoir with the sediments. Using the

initial ^{210}Pb activities at the time of formation (i.e., the time the reservoir was constructed) reliable estimates can be made to date the sediment (Appleby, 2001).

^{210}Pb is measured as supported ^{210}Pb which is derived from *in situ* decay of the parent radionuclide ^{226}Ra and unsupported ^{210}Pb which is derived from atmospheric flux. The supported ^{210}Pb will be in radioactive equilibrium with ^{226}Ra ; unsupported is determined by subtracting supported ^{210}Pb from total ^{210}Pb activity (Appleby, 2001).

The results show that total ^{210}Pb was significantly in excess of supporting ^{226}Ra in only the top 20 centimeters; however unsupported ^{210}Pb concentrations were very low. These results suggest the sedimentation rates have not been uniform during this period and it appears that the low ^{210}Pb concentrations are a result of dilution of atmospheric flux by rapid sedimentation. ^{137}Cs activity increased steadily with depth and reached its greatest value in the deepest section analyzed; sediment from 34-36 cm probably dates near the period of maximum fallout from the atmospheric testing of nuclear weapons, 1963. Assuming a mid-1906s date for this 34-36 cm section, the mean sedimentation rate during the past 40 years is approximately 0.95 centimeters per year. Since the cores that were collected contained no material at their bases, there was some uncertainty associated with estimated sedimentation rate for the Cottage Grove Reservoir.

Table 1: Summary of Mercury Concentrations Detected in Core Samples Collected in 2002

Depth Interval (centimeters)	Core #1			Core #2			Average Mercury Concentration (mg/kg-dry-wt)	Standard Error
	%Solids	Mercury Concentration (mg/kg-dry- wt)		%Solids	Mercury Concentration (mg/kg-dry- wt)			
0-2	20%	1.4	=	21%	1.4	=	1.4	0.027
2-4	27%	2.2	=	26%	1.6	=	1.9	0.38
4-6	36%	1.0	=	33%	1.0	=	1.0	0.0099
6-8	34%	0.88	=	34%	0.97	=	0.92	0.062
8-10	29%	0.84	=	31%	0.91	=	0.87	0.054
10-12	34%	0.79	=	34%	0.79	=	0.79	0.0019
12-14	34%	0.89	=	34%	0.93	=	0.91	0.034
14-16	39%	0.88	=	37%	1.1	=	1.00	0.17
16-18	39%	1.1	=	39%	0.96	=	1.0	0.068
18-20	38%	0.92	=	39%	0.85	=	0.88	0.054
20-22	40%	0.78	=	42%	0.89	=	0.83	0.077
22-24	45%	0.83	=	45%	0.81	=	0.82	0.012
26-28	44%	1.6	=	44%	1.4	=	1.5	0.085
28-30	44%	1.8	=	46%	1.8	=	1.8	0.0053
30-32	43%	2.4	=	43%	2.5	=	2.4	0.057
32-34	44%	3.9	=	46%	3.5	=	3.7	0.29
34-36	47%	2.3	=	46%	1.3	=	1.8	0.70
36-38	50%	2.4	=	43%	2.1	=	2.2	0.19

= - Indicates a detected concentration

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**Table 2: Summary of Mercury Concentrations
Detected in 1995 Core Sample**

Depth Interval (centimeters)	Mercury Concentration (mg/kg-dry- wt)
0-2	0.65 =
2-4	0.72 =
4-6	0.59 =
6-8	0.64 =
8-10	0.81 =
10-12	0.59 =
12-14	0.5 =
14-16	0.76 =
16-18	1.04 =
18-20	1.29 =
20-22	1.54 =
22-24	2.1 =
26-28	1.86 =

= - Indicates a detected concentration

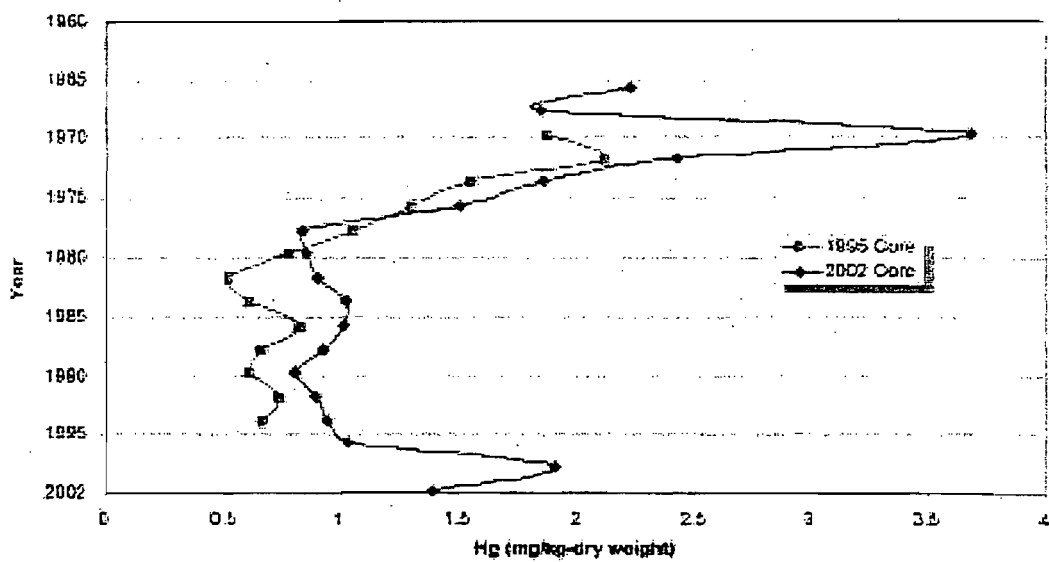


Figure 4: Profile of Mercury Concentrations in Core Samples

FOOD WEB SAMPLES

Mercury concentrations were measured in organisms representing three trophic levels: benthic invertebrates, omnivorous amphibians/fish, and piscivorous fish. Average mercury concentrations in Chironomid larvae and Anisoptera and Zygoptera nymphs (benthic invertebrates) were 0.049 mg/kg, 0.035 mg/kg, and 0.075 mg/kg, respectively (Fig. 5). Average mercury concentrations in fingerling catfish, snails, and tadpoles (omnivorous amphibians/fish) were 0.043 mg/kg, less than 0.017 mg/kg, and less than 0.021 mg/kg, respectively (Fig. 5). Mercury concentrations found in epaxial muscle tissue from largemouth bass ranged from 0.86 mg/kg to 1.6 mg/kg (Table 3).

Table 3: Summary of Mercury Concentrations in Epaxial Muscle from Largemouth Bass

Sample Type	Mercury Concentration (mg/kg-wet wt)	Sample Wt (g)
Largemouth bass	1.1 =	225
Largemouth bass	0.95 =	455
Largemouth bass	0.99 =	470
Largemouth bass	0.97 =	510
Largemouth bass	1.0 =	510
Largemouth bass	1.2 =	560
Largemouth bass	0.86 =	610
Largemouth bass	1.0 =	660
Largemouth bass	1.6 =	860
Largemouth bass	1.0 =	1700

= Indicates detected concentration

U Indicates concentration detected at the reporting limit

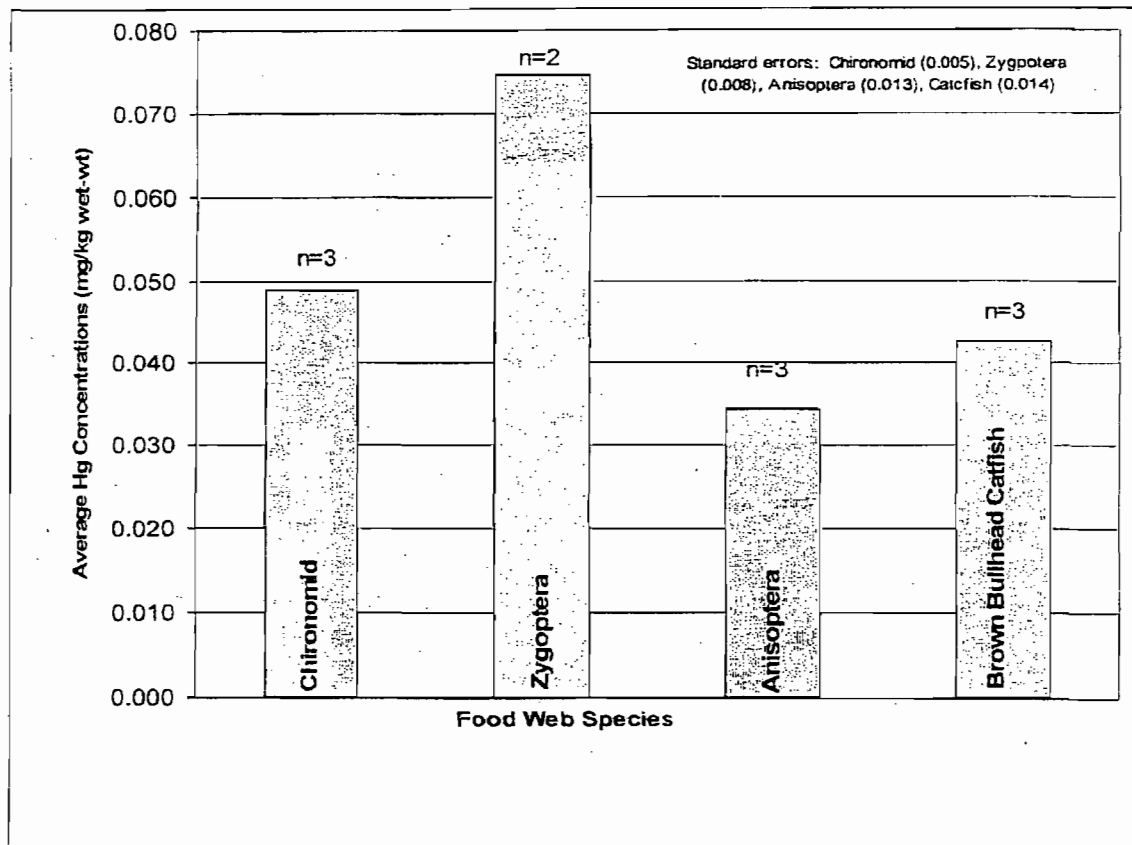


Figure 5: Average Mercury Concentrations in Food Web Samples

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DISCUSSION

MERCURY TRANSPORT WITHIN THE WATERSHED

Elevated mercury concentrations in soils at Black Butte Mine (190 mg/kg), surrounding the kiln (220 mg/kg to 270 mg/kg), and in the mine tailings (20 mg/kg) supports the conclusion that Black Butte Mine is a point source of contamination to the reservoir. As a result, mercury has entered the watershed from mining waste and probably atmospheric deposition of mercury vapor that escaped condensers during roasting of cinnabar. High soil mercury concentrations around the kiln support the role of atmospheric transport.

The chemical form of mercury affects transport in and between air, land, and water as well as chemical and biological behavior. The chemical forms of mercury that can undergo transformation includes elemental mercury [Hg(0)], inorganic mercury [Hg(II)], and methylmercury (MeHg) (Porcella, 1994). Some forms of mercury (e.g., Hg(II) or MeHg) may bind readily to organic molecules and to suspended solids that may be coated with an organic surface. When bound to suspended solids, the fate of these mercury forms is dependent upon deposition and erosion processes, and may eventually become a part of the sediment bed (Bale, 2000).

The percentage of sediment stored permanently by the reservoir and temporarily stored and then flushed through remains unknown. This reservoir has not been dredged in its 60 years history has been in existence. It is estimated that the annual sediment yield for the entire watershed is 11,040 cubic yards natural background and 1,442 cubic yards additional sediment from roads (BLM, 1997). Assuming equal distribution throughout the reservoir, it is estimated that it would take 500 years to add 3 feet of sediment to the reservoir, assuming permanent storage of this material (BLM, 1997). However, it is unlikely that uniform sediment distribution is occurring since the reservoir was constructed upon the original stream channel. It is likely that water flows more rapidly through this channel

than in other areas of the reservoir, resulting in scouring of the channel bottom and increasing the deposition rate in adjacent areas where the water flow is more gentle.

Mercury in tributary samples collected upstream of reservoir

Mercury concentrations in tributary stream sediments strongly correlate with its upstream or downstream proximity to the mine. Garoutte Creek, Dennis Creek, and Big River are three of the tributaries that form the Coast Fork of the Willamette River. Four samples were collected upgradient from the mine including, Little Creek, Brauti Creek, and two locations along Garoutte Creek. Concentrations of mercury in these samples range from 0.14 mg/kg to 0.46 mg/kg. It is possible that atmospheric transport of mercury in Kiln Fumes contributed to elevated concentrations in Little Creek and Brauti Creek compared to Garoutte Creek (Fig. 2).

Mercury concentrations from the samples collected along Big River were less than or equal to 0.020 mg/kg. The concentrations detected in the samples collected along Big River indicate that this tributary is not impacted by mining activities and are less than naturally occurring levels of mercury within this area. Khandoker (1997) reported naturally occurring levels of mercury in Cascade Range soils as < 0.09 mg/kg in A horizons and <0.05 mg/kg in B horizons. These concentrations are also less than the naturally occurring level of 0.11 mg/kg (for sediments <62 μm) established by the USGS indicating enrichment by natural processes or human activities (Rickert et al. 1977). Furthermore, mercury concentrations in the samples upgradient from the mine confirm the presence of naturally occurring ore deposits or atmospheric contamination, whereas samples collected along Big River show the absence of naturally occurring mercury or pollutant sources.

Mercury concentrations in sediments located downstream from the mine, in Dennis Creek and Garoutte Creek, range from 6.6 mg/kg (at the confluence of Dennis Creek and Garoutte Creek) to greater than 1.0 mg/kg at the confluence of Garoutte Creek with Big River. These results strongly suggest that the Mine area is a continuing source of mercury to these tributaries and also shows a strong concentration gradient from the area of the mine to the headwaters of the Coast Fork of the Willamette River.

Mercury concentrations in samples collected along the Coast Fork of the Willamette River range from 0.19 mg/kg to 1.4 mg/kg. These results suggest that some areas along the river have been scoured (i.e., sediment has been removed through erosion from select locations). Since these samples were collected during the summer, several months had lapsed since any significant storm events had occurred. During February 1995 (Park, 1996), sediment samples were collected from similar locations along the River. At this time, mercury concentrations ranged from 0.73 mg/kg to 1.3 mg/kg. During a storm event, erosion from the source area would increase, resulting in an increased deposition rate of mercury and a more significant mercury gradient from the source to the reservoir system, as shown in the results obtained during 1995.

Finally, mercury concentrations in samples collected from the creeks that flow directly into the reservoir (i.e., Wilson Creek, Cedar Creek, and Williams Creek) ranged from 0.03 mg/kg to 0.07 mg/kg. Concentrations observed in these samples indicate that the sediments have not been impacted by mining activities since concentrations are less than what is considered naturally occurring.

Mercury in tributary samples collected downstream of reservoir

Mercury concentrations in samples collected downstream from the reservoir increased with distance from the reservoir, with concentrations ranging from <0.02

mg/kg to 0.069 mg/kg. Without replicate samples robust statistical comparisons were unavailable. Similar to upstream locations, these results suggest that scouring has occurred. During February 1995 (Park, 1996), samples were collected from similar locations, however mercury concentrations decreased with distance from the reservoir. In this case when a storm event occurred, mercury stored in the sediment of the reservoir becomes scoured and is released through the dam resulting in higher mercury concentrations in downstream areas. Similar results were found in a study conducted at Lower Fox River in Wisconsin. High mercury concentrations in deeper river sediments coupled with scouring by periodic release of water through the DePere Dam operation transported high mercury concentrations downstream (Hurley et al. 1998).

MERCURY IN TRANSECT SAMPLES

Mercury concentrations in the surface sediment transect gradually increase from locations near the inlet to the dam. These results suggest an increased mercury deposition rate as suspended solids approach the dam. Deposition rates are likely to be influenced by increased flow rates from episodic hydrologic events as well as reservoir operation (i.e., annual water drawdown and drawdown from storm events).

Increased mercury concentrations are also likely to be associated with an increase in organic carbon content. Through visual inspection, organic carbon content of each transect sample increased from the inlet (where the reservoir bottom was primarily gravel or sandy material) to the spillway (where the reservoir bottom was primarily clay-type material). Organic carbon content or percent volatile solids (PVS) were not measured in the samples collected during 2002, however PVS was measured in the transect samples collected during 1995 (Park and Curtis, 1996). This study found increased mercury concentrations were associated with the clay-type sediments; however there was no correlation between PVS and the

mercury concentrations measured. The form of mercury likely to be found in the reservoir is likely to be associated with cinnabar (HgS). Since some forms of mercury readily bind to organic molecules and suspended solids with an organic surface, this provides another possible explanation for the gradual increase in mercury concentrations from the inlet to the spillway.

CORE SAMPLES/GEOCHRONOLOGY

Sample location strategy

Sediment cores were collected for the purpose of resolving temporal changes in mercury loading of the reservoir. This location strategy was selected because the deepest areas of a reservoir typically reflect the most stable regions and also have the highest sediment accumulation rates. Sediments in a lake are comprised of both allochthonous (external) and autochthonous (internal) inputs. To understand the temporal dynamics of the entire lake system, the core should be obtained from large integrative basins which blend internal inputs and external sources from all subcatchments. If cores are located in a flat region, it lessens the likelihood of erratic slumping of material to steep slopes. Finally, if cores are collected in deep regions of the reservoir, the sediment material is constantly submerged and less likely to be impacted by the effects of seasonal drawdowns, which would result in oxidation-reduction (redox) changes (Allen *et al.*, 1995).

Core dating methodology (^{210}Pb and ^{137}Cs)

The chronology of lake sediments can be determined by the presence of the natural radioactive isotope ^{210}Pb (half-life 22.3 years). The constant rate of supply method of dating is considered reliable in stable environments with uniform sediment accumulation rates. The ^{210}Pb chronology is independently verified by the presence of artificial radionuclides (i.e., ^{137}Cs). Fallout on a global scale began in 1954, and reached a peak in 1963 shortly after the test-ban treaty. However,

more recently fall-out from the Chernobyl reactor accident has been used to identify sediment deposited in 1986 (Appleby, 2001).

The core dating results show that total ^{210}Pb was significantly in excess of supporting ^{226}Ra in only the top 20 centimeters and unsupported ^{210}Pb concentrations were very low. These results suggest the sedimentation rates have not been uniform during this period and it appears that the low ^{210}Pb concentrations are a result of dilution of atmospheric flux by rapid sedimentation. ^{137}Cs activity increased steadily with depth and reached its greatest value in the deepest section analyzed; sediment from 34-36 cm was estimated to date from a period of maximum fallout from the atmospheric testing of nuclear weapons during 1963. Assuming a mid-1960's date for the 34-36 cm section, the mean sedimentation rate during the past 40 years is approximately 0.95 centimeters per year. Since the cores that were collected did not go to parent material, there is some uncertainty associated with estimated sedimentation rate for the Cottage Grove Reservoir.

Mercury concentrations of 2002 core

Average mercury concentrations found in the sediment core stratigraphy sample was shown in Figure 4. With the exception of an event that occurred from approximately 1969 to 1971, mercury input to the reservoir has gradually decreased from 1965 to 1979 (14 years). Since the reservoir was constructed in 1942, it is estimated that it has taken approximately 37 years for mercury input to stabilize. Thereafter, mercury input to the reservoir has remained relatively stable from 1979 to 2002.

Prior to construction of the reservoir, mercury was produced at the mine from approximately 1882-1926, and 1927 to 1943 (Brooks, 1973). After construction of the reservoir, mercury was periodically produced during 1951, 1957 to 1958, and

1965 to 1967. Mercury released into the watershed after the reservoir was constructed is likely related to the periodic mining activities that occurred through 1967 and is likely associated with the peak mercury concentration in 1971.

Although mercury production has ceased at the mine, mercury-rich mine waste and contaminated soil continues to serve as a source of mercury to the watershed.

As shown in the core profile, the reservoir serves as a sink for mercury released to the watershed above the reservoir, however it also serves as a source of mercury for release into the watershed below the reservoir.

The reservoir level is kept at summer high-pool elevation from late May to early September. Drawdown then begins so that winter low-pool elevation is reached by the end of October. When a storm occurs, water is held back in the reservoir and after the storm, the water is drained over a period of days or weeks to reach the appropriate level. From February to May, the reservoir is slowly filled to the summer high-pool elevation again (Ambers, 2001). Therefore, during spring and summer months the reservoir serves as a sink as a result of minimal water release. More detailed work during the fall and winter months is necessary to determine mercury loss from the reservoir with the increased release from operational activities and storm events.

FOOD WEB SAMPLES

Total mercury concentrations were measured in organisms representing three trophic levels, including benthic invertebrates, omnivorous amphibians/fish, and piscivorous fish (Figure 5, Table 3). Benthic invertebrate species include Chironomid larvae and Anisoptera and Zygoptera nymphs; omnivorous amphibians/fish species include snails, bullfrog tadpoles, and fingerling brown bullhead catfish; and piscivorous fish include largemouth bass.

Total mercury was measured in the foodweb samples where it was assumed that most mercury in the tissue was methylmercury. More than 90 percent of mercury in fish tissue (Allen-Gil et al. 1995) and more than 60 percent of mercury in benthic invertebrates (Wren et al. 1991; Fischer and Gustin, 2002) was methylmercury. The current study assessed contamination in the lower trophic levels of an aquatic foodweb, while earlier work focused on fish (Allen-Gill *et al.*, 1995; Park and Curtis, 1997).

In an aquatic foodweb, methylmercury is the most important form of mercury because it is highly bioavailable for uptake into aquatic organisms. Bioaccumulation of mercury into aquatic organisms can occur through multiple pathways including uptake from sediment and water, through the skin or cuticle, through ventilation of gills, and from consumption of contaminated sediment or prey (Post et al. 1996). The contribution from each pathway remains poorly understood and certainly species-dependent. Respiratory uptake has been identified as a substantial contributor to bioaccumulation, however the laboratory studies typically have involved aqueous concentrations that were orders of magnitude greater than those typically found in the field (Post et al. 1996). Field studies of mercury uptake in piscivorous fish have identified prey consumption as the primary uptake pathway (Grieb et al. 1990; Lindquist et al. 1991).

Biomagnification refers to the tendency of some chemicals to become increasingly concentrated at successively higher trophic levels of a food web. As a result, the larger and older fish have the highest amount of methylmercury in their tissues. A biomagnification factor (BMF) can be estimated when the concentration of mercury in organisms (or environmental media) at different trophic levels in a food chain are known and can be calculated as the ratio of the [Hg] in the predator (wet-weight)/ [Hg] in the prey (wet-weight). For the purpose of this study, the BMF for benthic invertebrates was estimated as the ratio of [Hg] in the benthic organism (wet-weight)/ [Hg] in sediment (wet-weight).

Methylmercury

Methylmercury is the most common form of organic mercury in the environment. In sediments, the production of methylmercury is favored under anoxic conditions and has been attributed primarily to sulfate reducing bacteria (Gilmore and Henry, 1991; Gilmore et al. 1992). Abiotic methylmercury production in natural environments has been shown to be of minor importance (Berman and Bartha, 1986).

Past and current studies have been conducted to determine the environmental conditions that favor or suppress the formation of methylmercury in the aquatic environment. Results suggest that sulfate reducing bacteria are important for mercury methylation, and their methylating activity is influenced by the concentration of sulfate in the surrounding environment (Chen et al. 1997). However, a more recent study suggests that factors influencing microbial methylmercury production includes microbial community composition, mercury availability, carbon availability, and the abundance of electron acceptors such as sulfate (Macalady et al. 2000). This study also suggests that other bacterial groups, in addition to sulfate reducing bacteria, are of potential importance for methylmercury production.

Recent studies have shown that methylmercury production in sediment can be reduced or inhibited as a result of controlling various water quality parameters. A study within the Carson River-Lahontan Reservoir system found that the rate of methylmercury production was reduced by increasing pH and methylmercury production was inhibited by the presence of group VI anions (Bonzongo et al. 1996; Chen et al. 1997).

A study comparing the availability of tracer and ambient mercury was conducted to determine whether mercury methylation or demethylation controls the levels of methylmercury in the aquatic environment (Hintelmann et al. 2000). This study found that methylmercury levels in sediment were controlled by both methylation

and demethylation, and the relative importance of each reaction is likely dependent upon environmental conditions and biological factors with spatial and temporal variations. An estimated sediment half-life of less than two days for methylmercury, suggests it is not persistent in aquatic systems and a constant supply of methylmercury is necessary to maintain steady-state concentrations. Possible demethylation end-products includes the formation of divalent mercury through oxidative demethylation, the formation of elemental mercury through a reductive process, or mercury volatilization.

Benthic invertebrates

The benthic invertebrates collected for this study live in direct contact with surficial sediment and detritus (Chironomid larvae, and Zygoptera and Anisoptera nymphs). Chironomid larvae were collected from surface sediment near the spillway of the reservoir at approximately 16 meters depth during July 2002. The Chironomid lifecycle is variable; some forms have only one generation in two years, whereas others have several generations in a single year (Pennak, 1978). Chironomid larvae are primarily herbivorous and feed on algae, higher aquatic plants, and organic detritus; however it is likely that coincidental sediment absorption occurs through bulk processing (Pennak, 1978). The average mercury concentration measured in the Chironomid larvae was 0.049 mg/kg (Figure 5).

This suggested a relationship between sediment mercury concentrations and the body burden of mercury. The mercury analysis performed could not distinguish between the forms of mercury (methyl or total) or whether the mercury originated from the tissue or the contents of the gut. Therefore it is difficult to determine if the mercury measured in these organisms is the result of sediment uptake (i.e. gut contents) or uptake across the skin. Uptake from respiration through the skin could occur since these samples were collected in an anaerobic environment under reducing conditions, which is considered conducive for mercury methylation.

As described earlier, mercury concentrations were measured in surface sediment in the vicinity of Chironomid collection from the reservoir. The BMF from sediment (1.4 mg/kg wet-weight) to the Chironomid larvae is equal to 0.035. This suggested little accumulation of mercury from sediment by this species.

Zygoptera and Anisoptera nymphs were collected from locations near the inlet of the reservoir at water depths of about one meter during August 2002. It is probable that the lifecycle for the great majority of these species includes 11 to 14 instars. The length of each instar is dependent upon the species and the prevailing temperature and food conditions (Pennak, 1978). The Zygoptera nymphs can be distinguished from Anisoptera nymphs by the presence of three leaflike tracheal gills at the top of the abdomen. Food consists primarily of other aquatic insects, annelids, and small Crustacea and mollusks (Pennak, 1978). The average mercury concentrations in Zygoptera and Anisoptera nymphs were 0.075 mg/kg and 0.034 mg/kg, respectively (Fig. 5). The mercury measured in these organisms could be the result of uptake across the cuticle and/or gills, and through consumption of prey. The mercury concentrations measured in the Zygoptera nymphs were approximately twice the concentration measured in the Anisoptera nymphs, suggesting that the Zygoptera nymph may be a more voracious predator.

The mercury concentration in sediment collected from a similar location (surface sediment transect sample) was 0.55 mg/kg wet-weight. The BMF from sediment to the Zygoptera and Anisoptera nymphs is equal to 0.14 and 0.06, respectively, suggesting no biomagnification at this trophic level.

Omnivorous/herbivorous amphibians and fish

Snails, tadpoles, and fingerling brown bullhead catfish were collected from the vegetative layer near the inlet of the reservoir at water depths of about 1 meter during August 2002.

The lifecycle for the majority of snail species ranges from nine to 15 months. Snails are considered vegetarians and feed primarily on algae and dead plant material (Pennak, 1978). For bullfrog tadpoles, the aquatic phase of their lifecycle is approximately three months from the egg until metamorphosis is complete. Tadpoles feed primarily on algae and microorganisms suspended in water (Storer et al. 1979). Mercury was not present at detectable concentrations in any of the snails or tadpoles analyzed (less than 0.02 mg/kg). The absence of mercury is considered reasonable because the pathways for bioaccumulation of mercury are likely incomplete. These species were obtained from the vegetative layer and do not live in direct contact with the sediment; they are herbivores and do not consume prey, and finally mercury measured in water collected during a previous study at the reservoir was present at very low concentrations (0.78 $\mu\text{g/L}$) (Allen-Gil et al. 1995). Consumption of these species as prey likely does not contribute to biomagnification in higher trophic levels.

The fingerling brown bullhead catfish are bottom-feeding omnivores; they are primarily herbivorous, however they can be predaceous and feed on small aquatic animals including fishes (Storer et al. 1979). The average mercury concentration measured in the fingerling catfish was 0.043 mg/kg (Fig. 5). The mercury concentration measured in Zygoptera nymphs collected from a similar location was 0.075 mg/kg. The BMF from the Zygoptera nymph to the fingerling catfish was 0.57, suggesting potential for biomagnification at this stage in the catfish lifecycle.

Piscivorous fish

Mercury concentrations were measured in epaxial muscle from 10 largemouth bass collected in 1998 by Oregon DEQ (unpublished data). Mercury concentrations were measured in fillets (no skin or rib bones) from the dorsal to the belly and between the pectoral and dorsal fins. Mercury concentrations ranged

from 0.86 mg/kg to 1.6 mg/kg² (Table 3). The age, length, and time of year these samples were collected were not provided.

In largemouth bass collected between 1990 and 1992 (Allen-Gil *et al.*, 1995), mercury concentrations in largemouth bass ranged from approximately 0.5 mg/kg at three years of age to greater than 1.5 mg/kg in fish at five years of age. In largemouth bass collected between 1993 and 1995 (Park and Curtis 1997), mercury concentrations in largemouth bass ranged from approximately 0.5 mg/kg at three years to approximately 0.7 mg/kg in fish at five years of age. Largemouth bass collected during 1990 and 1992 showed a positive correlation between fish age and mercury content, with little or no bioaccumulation of mercury occurring between one and three years of age, followed by a linear increase thereafter. This correlation was not observed in the Park and Curtis (1997) study. Although, age and length were not provided for fish in this study, based on the reported concentrations it is likely that these fish are older than 3 years of age. If the fish tissue collected during 1998 were of comparable age to the two previous studies, the concentrations of methylmercury indicate concentrations have increased.

Largemouth bass are fish-eating predators, although their diet also includes invertebrates and amphibians. Mercury concentrations were measured in bluegill sunfish collected from the Cottage Grove Reservoir (Park and Curtis 1997). Mercury concentrations were 0.43 mg/kg (2 years of age), 0.63 mg/kg (3 years of age), 0.45 mg/kg (4 years of age), and 1.1 mg/kg (5 years of age). Bluegill sunfish are a likely prey item for largemouth bass and was used to estimate a BMF for this trophic level. Using the mercury concentration of 0.63 mg/kg for the bluegill sunfish and an average mercury concentration of 1.1 mg/kg in largemouth bass, the BMF at this level is approximately 1.7. These results suggest that significant biomagnification is occurring at the higher trophic levels.

Factors influencing bioaccumulation

In a previous study at Cottage Grove reservoir, (Allen-Gil et al. 1995) reported four broad categories that favorably influence mercury bioaccumulation. These include hydrologic factors, water chemistry, sediment characteristics, and life history of the fish.

Hydrologic factors that favorably influence bioaccumulation include slow flow, frequent flooding and recent impoundment of a reservoir (Allen-Gil et al. 1995). It is unlikely that bioaccumulation occurring at the reservoir is influenced by the hydrologic factors of the reservoir. Sixty years have lapsed since this reservoir was constructed, thereby precluding recent impoundment as a contributing factor. This reservoir is seasonally managed where water flow and flooding is seasonally influenced by management practices.

Water chemistry parameters that favorably influence bioaccumulation include low conductivity, high dissolved organic content, a pH less than 6.0 or greater than 8.5, and high temperature (Allen-Gil et al. 1995). The water chemistry parameters measured at the reservoir in September 1989 do not suggest that bioaccumulation would be favorably influenced. At that time, conductivity was relatively low (56 μ mhos), however pH was not within the favorable range (pH 7.7), and dissolved organic carbon (DOC) content was not measured. Most studies show a fairly consistent negative correlation between fish mercury content and pH, alkalinity, calcium, and conductivity (Grieb et al. 1990). Grieb et al. (1990) also showed that DOC and mercury concentrations did not correlate in drainage lakes and showed a consistent and statistically significant negative correlation in seepage lakes.

Twelve water samples from the reservoir were analyzed for mercury reporting a mean concentration of 0.78 μ g/L (Allen-Gil et al. 1995). This study concluded that mercury was likely to be associated with the particulate fraction, and that it may

² All mercury concentrations in aquatic food web samples are reported on a wet-weight basis.

not be as readily absorbed in biota as dissolved inorganic mercury or methylmercury (Stokes and Wren, 1987).

Sediment characteristics that favorably influence bioaccumulation include a mildly oxidizing environment, low clay content, high organic content, and low levels of complexing agents (Allen-Gil et al. 1995). Sediment collected from the shoreline of the reservoir between 1989 and 1992 was classified as sand-sandy loam, with an average clay content of 10 percent, an average carbon content of 7.1 percent, and approximately 33 percent of total mercury was associated with fine grain (Allen-Gil et al. 1995). In the sediment collected from the reservoir in 1994 (Park 1996), sediment mercury concentrations were not significantly correlated with organic content (i.e., percent volatile solids). These samples differ in composition from the (Allen-Gil et al. 1995) samples because they were collected from deeper waters where the organic content was significantly higher.

Life history characteristics of fish that favorably influence bioaccumulation include large size, long life span, and a high trophic position in the food web (Allen-Gil et al. 1995). Largemouth bass are considered long-lived and have the largest body sizes and probably the lowest rates of growth and metabolism at older ages (Scott et al. 1973). Life history is likely the strongest factor influencing bioaccumulation at Cottage Grove Reservoir.

In a recent study (Rose et al. 1999), largemouth bass were collected from 24 lakes not likely to have been impacted by non-point sources. These lakes were selected to evaluate the importance of ecoregional differences. This study showed that mercury concentrations in bass were most strongly and positively associated with the weight of the fish, lake size, and variables representing potential source area-contribution sizes (wetlands and watersheds). It concluded that considerable variation associated with size or food-chain position in largemouth bass tends to obscure relationships between environmental variables and mercury bioaccumulation.

In a study by Post (1996), the proportion of mercury uptake across the gills and from food consumption reflect seasonal patterns in temperature, body size dependent energetics, and diet. When growth rates and temperatures were low, in the spring and fall, the largest proportion of mercury uptake was across the gill. In the summer, when temperature and consumption rates were the highest, the proportion of mercury uptake from consumption exceeded the uptake across the gill. This study illustrates the importance of mercury concentration and speciation in the water and food. If steady-state is established between mercury concentrations in water and food organisms, then the proportional uptake rates are less affected by overall environmental mercury levels, and more by the diet of the fish. This study indicates that the bioavailability of mercury in sediment is unclear, and the emphasis on mercury contamination by large-bodied piscivorous fish has hindered the ability to understand the links between elemental sources of mercury and the complex direct and indirect food web processes.

Health effects from exposure to methylmercury

The primary exposure route for humans to methylmercury is from consumption of contaminated fish or marine mammals. Methylmercury is easily absorbed by the gastrointestinal tract, where it enters the bloodstream and becomes transported to other parts of the body. Methylmercury in the bloodstream can pass through the blood-brain barrier; it can also be transported from the blood of pregnant women into the blood of the developing child and then into the child's brain and tissue. Methylmercury can also be excreted into breast milk resulting in exposure of the nursing child. Children are considered more sensitive to methylmercury than adults because it can easily pass into the developing brain of a young child which may interfere with the development process. Exposure of children may result in small decreases in IQ if the exposure was small, to more severe effects including brain damage with mental retardation, incoordination, or the inability to move.

The most severe effects include blindness, involuntary muscle contractions and seizures, muscle weakness, and the inability to speak. The most extreme cases of neurotoxicity are associated with the ingestion of fish containing methylmercury in the Minimata area of Japan and from ingestion of bread made from wheat and other cereals treated with methylmercury fungicide. However, current research is being conducted on the health effects from low-level exposures (ATSDR 2000).

With the exception of one sample, all concentrations measured in largemouth bass were above the FDA action limit of 1.0 mg/kg for commercially-caught fish. All concentrations are greater than the Oregon Department of Human Services action limit of 0.35 mg/kg. A fish consumption advisory has also been established by the Oregon Department of Human Services (published in 1993) specifically for fish caught at the Cottage Grove Reservoir. This advisory recommends that pregnant women, nursing women, and children up to six years of age should not consume any fish from the reservoir; and children older than six years and healthy adults should limit their consumption of fish from this reservoir to no more than one-half pound (8 ounces) of fish per week. Public health advisories are issued to help prevent noncommercial fishermen and their families from consuming fish containing mercury.

CONCLUSIONS

This study accomplished the general objective of confirming a point source and surveying the distribution of mercury contamination in the basin of Cottage Grove Reservoir. The objectives were accomplished through comparing the mercury stratigraphy in the 1995 and 2002 sediment core samples, estimating the deposition rates and long-term trends for sediment accumulation in the reservoir, and assessing the level of mercury contamination in the lower trophic levels of an aquatic food web.

Elevated mercury concentrations in soils surrounding the Black Butte Mine supports the conclusion that the Black Butte Mine is a point source of contamination to the reservoir. Mercury concentrations observed in the tributary surface sediment samples indicate that Dennis Creek, Garoutte Creek, and the Coast Fork of the Willamette River (upstream and downstream from the reservoir) continue to be influenced by the mine site. Mercury concentrations found in locations upgradient from the mine are similar to areas not disturbed by mining. Big River and the creeks that feed directly into the reservoir contain sediments with very low mercury concentrations and indicate they have not been influenced by past mining activities. Concentrations of mercury observed in the contaminated tributaries are heavily influenced by storm events and reservoir management practices. After a storm event, erosion from the source area increases, resulting in increased deposition of mercury in downstream areas.

Mercury concentrations in surface sediment transect samples gradually increase from the reservoir inlet to the spillway, suggesting an increased mercury deposition rate as suspended solids approach the dam. The geochronology of the sediment core stratigraphy samples indicate mercury input to the reservoir has remained relatively constant over the past 20 years; however the mine acts as a continuing source. Core dating results indicate that the sediment deposition rate

is approximately 0.95 cm per year. It is unlikely that mercury input will further decrease unless the source of the mercury is contained (that is, the mine tailings). Following containment, new sediments eventually bury the old deposits leading to recovery, but mixing and entrainment of sediments by bioturbation and episodic resuspension can significantly retard recovery (Bale, 2000).

Finally, the level of mercury contamination in lower trophic levels was assessed. Mercury concentrations in benthic organisms and the omnivorous/herbivorous amphibians and fish suggest little biomagnification at these lower trophic levels when compared to mercury concentrations in sediment or prey. However, concentrations of mercury in largemouth bass indicate that significant biomagnification is occurring within the food web. Concentrations of mercury measured in the fish tissue were greater than the Oregon Department of Human Service action level of 0.35 mg/kg and also greater than the FDA action limit of 1.0 mg/kg for commercially caught fish. The concentrations presented in this study are slightly greater than the concentrations measured in fish tissue from previous studies. It appears that the most important factor influencing bioaccumulation in fish tissue is the life history of the fish species. Although bioaccumulation can occur through multiple pathways, it appears that diet may be most important.

The remediation of mercury contaminated sites may depend upon gaining an understanding of the factors that make mercury bioavailable and mobile. This could include determining the species of mercury (i.e., HgS, Hg(0), or Hg(II)) at the reservoir that are available for uptake. In addition, future studies may need to focus on lower trophic levels to better evaluate the effects of environmental variables and understand the links between sources of mercury available for uptake and the complex direct and indirect food web processes.

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